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# THE CRITICALITY OF HETEROGENEOUS LATTICES OF EXPERIMENTAL BERYLLIUM OXIDE REACTOR FUEL PINS IN WATER AND IN AQUEOUS SOLUTIONS CONTAINING BORON AND URANYL NITRATE

E. B. Johnson

OAK RIDGE NATIONAL LABORATORY
OPERATED BY UNION CARBIDE CORPORATION FOR THE ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION

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# THE CRITICALITY OF HETEROGENEOUS LATTICES OF EXPERIMENTAL BERYLLIUM OXIDE REACTOR FUEL PINS IN WATER AND IN AQUEOUS SOLUTIONS CONTAINING BORON AND URANYL NITRATE

E. B. Johnson

JULY 1976

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ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION

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# THE CRITICALITY OF HETEROGENEOUS LATTICES OF EXPERIMENTAL BERYLLIUM OXIDE REACTOR FUEL PINS IN WATER AND IN AQUEOUS SOLUTIONS CONTAINING BORON AND URANYL NITRATE

E. B. Johnson

## ABSTRACT

The fuel intended for the Experimental Beryllium Oxide Reactor was made available for a series of critical experiments at the Oak Ridge Critical Experiments Facility. The fuel pins consisted of compressed ceramic pellets contained in Hastelloy X-280 tubes. The pellets were a homogeneous mixture of $U(62.4)O_2$ and BeO. The data generated are applicable to support the establishment of specifications for the chemical recovery of uranium in a process in which boron as a soluble neutron absorber is a part of the dissolver solution. It was found that a boron concentration of about 0.3 g/liter in a dilute solution of uranyl nitrate (~3.7 g of $^{235}U$/liter) increased the critical mass 60% over that without boron.

## INTRODUCTION

The Experimental Beryllium Oxide Reactor (EBOR) was designed in the early 1960's by the General Atomic Division of General Dynamics Corporation. It was to have been operated at the National Reactor Testing Station (NRTS) at a power of 10 MW thermal as a beryllium oxide-moderated and -reflected helium-cooled reactor with high neutron leakage and an epithermal-energy neutron spectrum. The program was terminated prior to fuel loading and the uranium in the fabricated elements was scheduled for recovery by the Idaho Chemical Processing Plant (ICPP). The absence of criticality data for uranium of any enrichment homogeneously mixed with beryllium on which to base chemical recovery specifications prompted transfer of the fabricated fuel elements to the Oak Ridge Critical Experiments Facility (CEF) for partial disassembly in order to obtain the fuel pins for a limited series of critical experiments in support of projected Chemical Plant operations. The experimental program was supported by the ICCP.

In chemical recovery of uranium from spent fuel, the material is put into solution in a dissolver. For reasons of economics the charge into the dissolver should be as large as possible consistent with nuclear criticality safety. In order to increase the permissible size of a charge, some chemical plants[1] are using boron as a soluble neutron absorber in the dissolver solution. Experiments have been reported[2,3] in which boron was added to the moderator-reflector water of lattices of fuel elements to determine the increase in critical mass. The present investigation was an extension of those measurements to fuel containing uranium enriched to 62.4% $^{235}U$ and diluted with BeO.

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1. W. G. Morrison, Criticality Aspects of the Revised Zirconium Dissolution System at the Idaho Chemical Processing Plant, Idaho Nuclear Corporation Report IN-1173 (February 1968).
2. E. B. Johnson and R. K. Reedy, Critical Experiments with SPERT-D Fuel Elements, Oak Ridge National Laboratory Report ORNL-TM-1207 (July 1965).
3. E. B. Johnson, Critical Lattices of U(4.89) Rods in Water and in Aqueous Boron Solution, Trans. Am. Nucl. Soc., 11, 674 (1968).

## DESCRIPTION OF FUEL

The fuel was contained in compacted ceramic pellets which were a homogeneous mixture of $UO_2$ and BeO. Each fuel pin consisted of a stacked column of fuel pellets in Hastelloy X-280 tubing. The pellets and pins are described in Table 1. After the pellets were installed in the tubes, the clearance between pellets and tubing was eliminated by creep-shrinking. Since each pellet had a shallow circumferential groove at its midplane into which the tubing was shrunk, its vertical position in the pin was fixed. Figure 1 gives the dimensions of the pellets and of their assembly in the cladding. A thin, narrow helical spacer was projection-welded every half-inch to the outside of the cladding tube and could not be conveniently removed. Their presence introduced some non-uniformity in the spacing of the fuel pins in lattices.

Figure 2 shows a cross section of a fuel element. The core was a cylinder of ceramic BeO around which were arranged, equally spaced in a circle, 18 fuel pins; Fig. 3 is a photograph of a portion of the top of a partially disassembled element showing this arrangement. The helical spacers mentioned above and the tubing shrunk into the pellet midplane grooves are visible. Surrounding the fuel pins was a shroud tube of Hastelloy X-280 that served as the outer boundary for the helium coolant flow and directed the flow around the fuel pins and the central spine. Spacing between the fuel tubes and the central spine and the shroud tube was maintained by the helices. The outer boundary of the fuel element was defined by square annular BeO moderator-reflector blocks. A stack of 59 of these blocks (a total height of 88.4 in.) surrounded the shroud tube; each block in the stack was joined to the adjacent blocks by BeO dowel pins located at diagonally opposite corners in order to provide torsional and lateral rigidity. The Be:$^{235}$U atomic ratio in the assembled element was[4] 117. Each element contained 2.81 kg of $^{235}$U.

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4. Experimental Beryllium Oxide Reactor Program. Quarterly Progress Report for the Period Ending March 31, 1964, General Atomics Report GA-5238 (April 30, 1964).

Analysis of the BeO showed the following impurities, their abundance expressed in parts per million:

| | | | | | |
|---|---|---|---|---|---|
| Ag | 5 | Cu | 4 | Mn | 7 |
| Cr | 7 | Mg | 1440 | V | 35 |
| Ni | 4 | Ti | 55 | Ba | 5 |
| Al | 3600 | Nb | 35 | Si | 3600 |
| C | 82 | Fe | 290 | | |

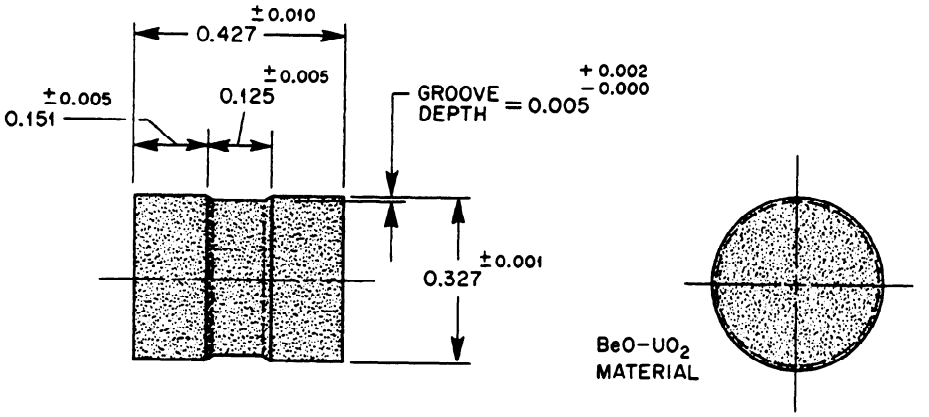
It was established that the beryllium removable from the surfaces by wiping was insufficient to be a personnel hazard of concern as an ingested poison.

Table 1. Fuel pins from the EBOR elements

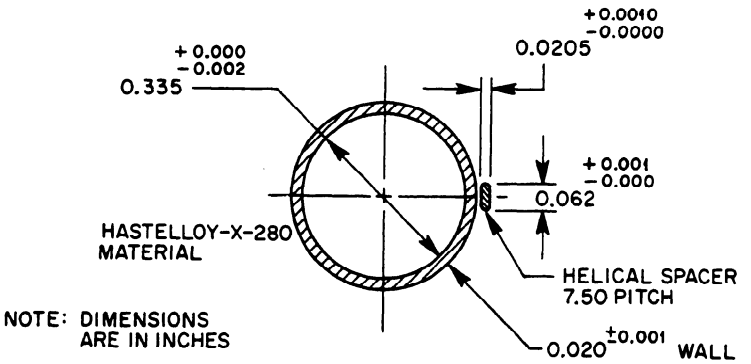
| | |
|---|---|
| Isotopic composition of uranium[5] (wt %): | |
| $^{234}U$ | 0.42 |
| $^{235}U$ | 62.4 |
| $^{236}U$ | 0.29 |
| $^{238}U$ | 36.9 |
| Fuel pellet: | |
| Diameter | 0.327 in. (0.831 cm) |
| Height | 0.427 in. (1.085 cm) |
| Fuel length in tube: | 76 in. (193 cm) |
| Hastelloy X-280 cladding: | |
| Outside diameter | 0.375 in. (0.952 cm) |
| Inside diameter | 0.020 in. (0.051 cm) |
| Helix (on outer surface of clad): | |
| Width | 0.062 in. (0.157 cm) |
| Thickness | 0.020 in. (0.051 cm) |
| Pitch | 7.5 in. (19.05 cm) |
| Fuel composition: | |
| $U(62.4)O_2$-BeO homogeneous ceramic | |
| 50.2 wt % uranium | |
| 43 wt % beryllium oxide | |
| Minimum density: 94% of theoretical | |
| $^{235}U$ (average per pin determined from transfer data for those elements used in experiments): | 156.27 g |
| Be:$^{235}U$ atomic ratio: | 13 |

5. A. D. McWhirter and A. J. Goodjohn, The Physics Characteristics of the Experimental Beryllium Oxide Reactor, General Atomics Report GA-4113 (October 24, 1963).

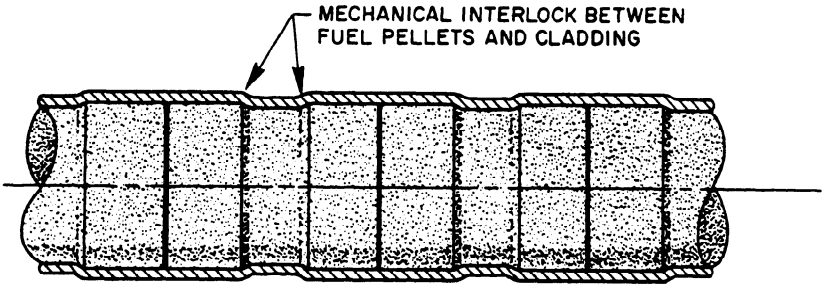
ORNL-DWG 75-6698




(*a*) FUEL PELLET




(*b*) FUEL CLADDING




(*c*) FUEL PELLET–FUEL CLADDING ASSEMBLY AFTER CREEP–SHRINK PROCESS

Fig. 1. Fuel pellet and assembly in cladding

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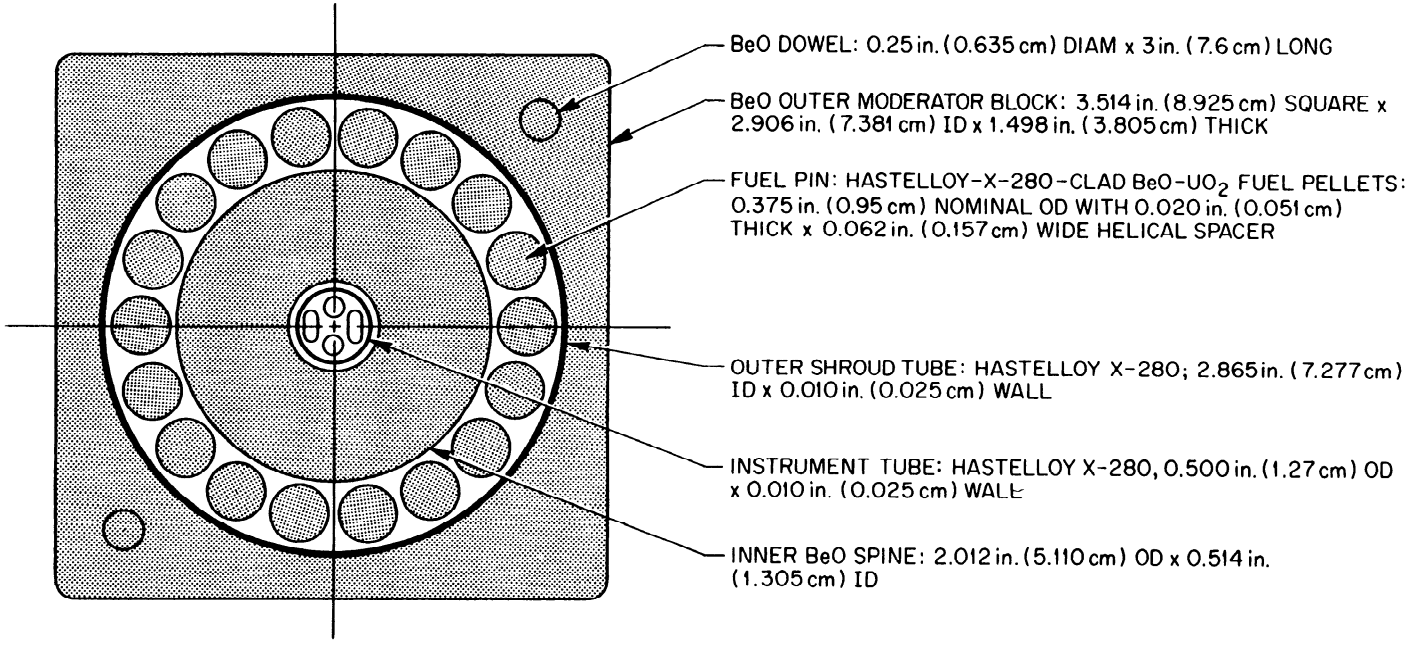


Fig. 2. Cross section of the fueled region of an EBOR fuel element

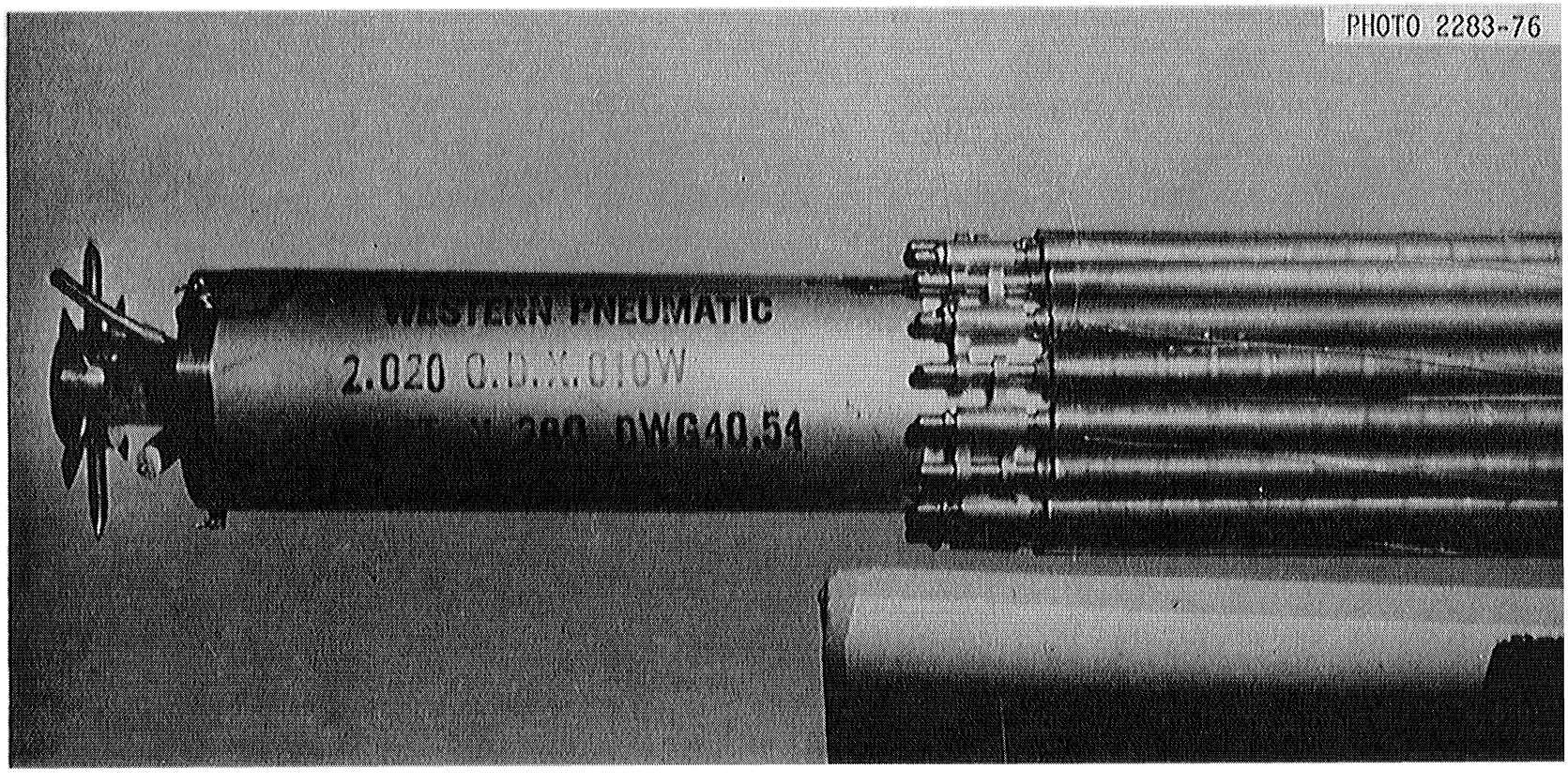


Fig. 3. The top of a partially disassembled EBOR fuel element

## EXPERIMENTS

### Fuel Elements

The fuel elements were received in the shipping casks in which they had been transported from the manufacturer to NRTS. Since it was necessary to store the elements outside the shipping casks at the CEF until the fuel pins could be removed, it was necessary to establish an arrangement that would be subcritical. Guidance for the disassembly operation had been supplied by General Atomics[6] and included some of the criticality safety rules enforced during their fabrication. Relevant to CEF operations were the criteria that a separation of 6 ft be maintained between elements in a planar array and that the pins also be arranged on a plane with a center spacing no less than 1.5 in. Since the space necessary to meet these criteria was not readily available at the CEF, subcritical storage arrangements were established directly by critical experiments.

It was recognized that the elements were well moderated (Be:$^{235}$U atomic ratio = 117, considering all the beryllium in the element) and that reflection, as by concrete floors, could make an appreciable contribution to reactivity. Therefore, as a first step, a single element was installed in a 9-ft-diam stainless-steel-lined tank to which water could be added by remote operation. The element was supported essentially horizontally on a Plexiglas frame so that more than a 6-in. thickness of water could serve as reflector between the element and the bottom of the tank. Addition of an effectively infinite water reflector produced no measurable increase in neutron multiplication. A total of 10 submerged elements in contact in a single plane, added one at a time and the resulting array completely reflected by water, had no detected neutron multiplication. This array, shown in Fig. 4, had the largest area that could be accomodated in the tank. An array of 16 elements in two 8-element layers in contact, also assembled in steps, was subcritical. Table 2 shows the results of the few additional experiments,

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6. Letter from B. Turvovlin to H. N. Wellhouser, Procedure for Disassembly of EBOR Fuel Elements, dated February 15, 1967.

some of which were critical, that were done with the fuel elements themselves. No attempt was made to extend the measurements to arrays of more than two tiers because of the limited intended use for the data. As a result of these experiments, the elements were stored in a single layer on concrete at spacings convenient for handling in an area where additional significant reflectors could not exist.

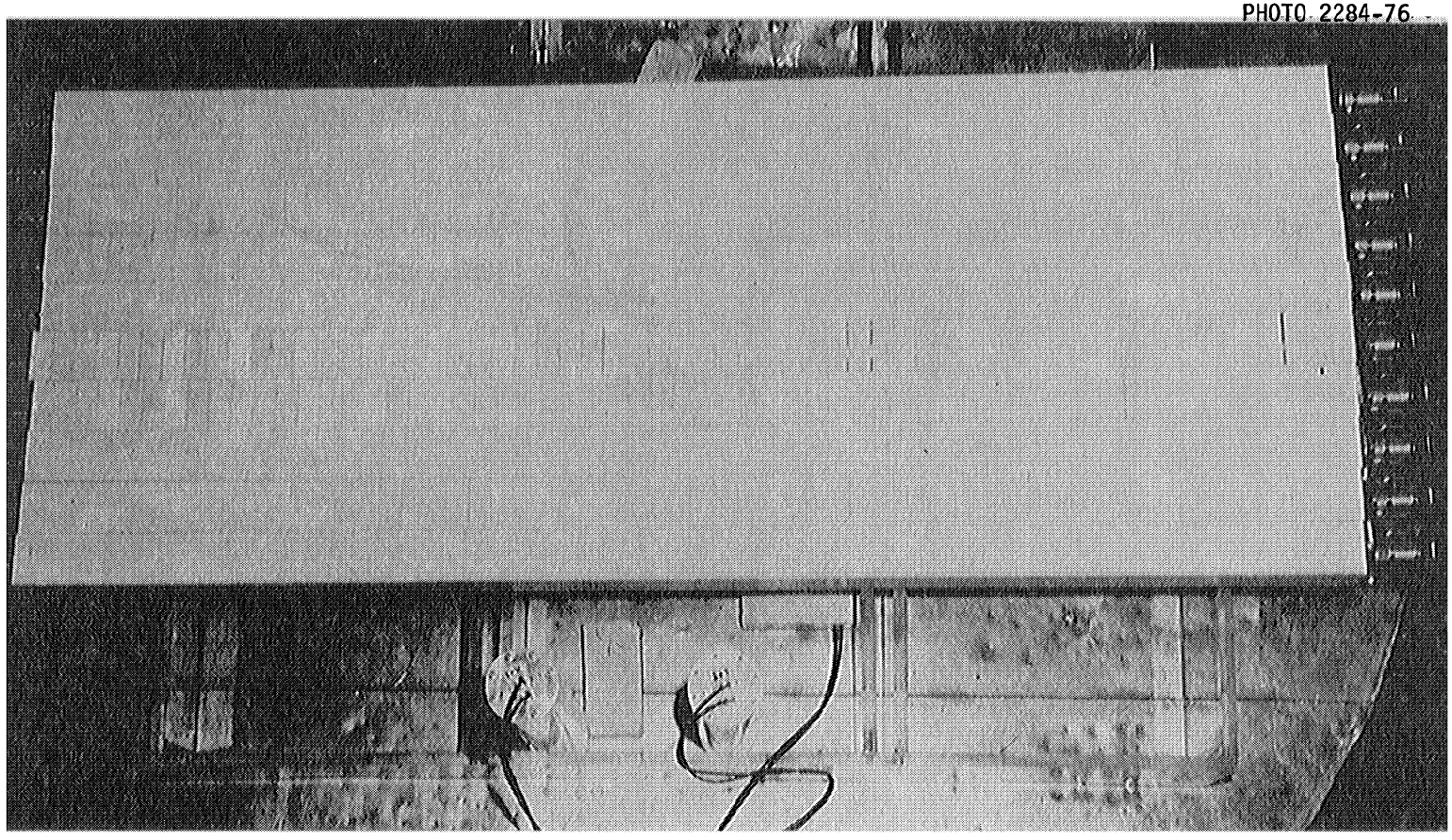


Fig. 4. A slab lattice of EBOR fuel elements. This lattice was subcritical when submerged.

Table 2. Lattices of EBOR fuel elements in water

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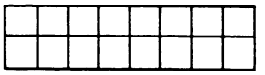
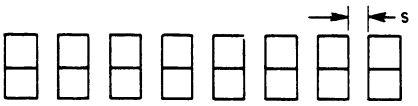
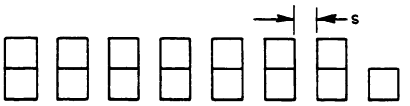
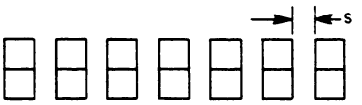
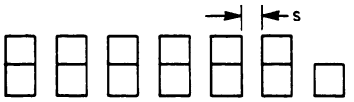
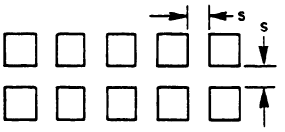
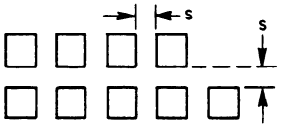
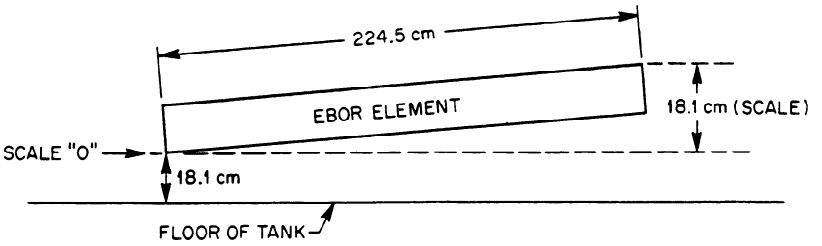
| ARRANGEMENT OF ELEMENTS (ELEVATION) | SURFACE SPACING (in.) | SURFACE SPACING (cm) | CRITICAL WATER HEIGHT[a] (cm) |
|---|---|---|---|
|  | 0.00 | 0.00 | SUBCRITICAL |
|   | 0.75 | 1.90 | 28.6 |
|   | 0.75 | 1.90 | 31.1 |
|   | 0.75<br>0.50 | 1.90<br>1.27 | SUBCRITICAL<br>31.0 |
|   | 0.50<br>0.25 | 1.27<br>0.63 | SUBCRITICAL<br>SUBCRITICAL |
|   | 0.50 | 1.27 | 31.0 |
|   | 0.50 | 1.27 | SUBCRITICAL |

a. THE WATER HEIGHT QUOTED HERE IS THAT MEASURED FROM THE BOTTOM OF THE LOWEST SURFACE OF THE OUTER BeO BLOCKS, i.e., FROM THE "0" REFERENCE INDICATED IN THE DIAGRAM BELOW.



Fuel Pins in Water

After the fuel pins had been removed from the elements, they were latticed in water at a number of spacings in order to determine the spacing at which the critical mass was minimal. The separation between pins was established by Plexiglas spacers located at three elevations. Aluminum Unistrut, also at three elevations, provided lateral support and stability for the lattices and maintained the desired spacings. Figure 5 shows a lattice of pins arranged in square pattern; this lattice was critical when submerged. Figure 6 is a photograph of the only lattice assembled in triangular pattern. In each case provision was made for at least a 6-in.-thick bottom water reflector.

The data for lattices moderated and reflected by water are summarized in Table 3 and plotted in Fig. 7. The separation of pins was determined from several, usually at least 10, measurements of the outside dimensions of each array at as many different locations and the outside diameter of the fuel cladding tube. The Plexiglas spacer thicknesses were between 0.100 and 0.800 in., at 0.1-in. intervals, and the spacers within a set were uniform. The average surface separations actually established differed from precise tenths of an inch by amounts between 0.004 and 0.021 in., which are the result of the random orientation of the helices with respect to the Plexiglas spacers.

The first entry in Table 3 is a subcritical close-packed lattice of 59.4 kg of $^{235}U$ in 380 fuel pins; this lattice is also indicated on Fig. 7. The surface separation of the pins in this lattice averaged 0.040 in. (0.102 cm), twice the thickness of the helices, rather than zero; because of these spacers, attached to the fuel pins, actual contact between adjacent pins could not be established.

Two lattices at a nominal surface separation of 0.5 in. were assembled (Numbers 6 and 7 of Table 3) in order to determine whether lattice spacings were readily reproducible and to investigate the effect of array shape on criticality. The average surface spacing in the two lattices differed by 0.009 in. and a slightly higher critical mass was observed for the wider spacing which, considering the fact that the

PHOTO 2285-76

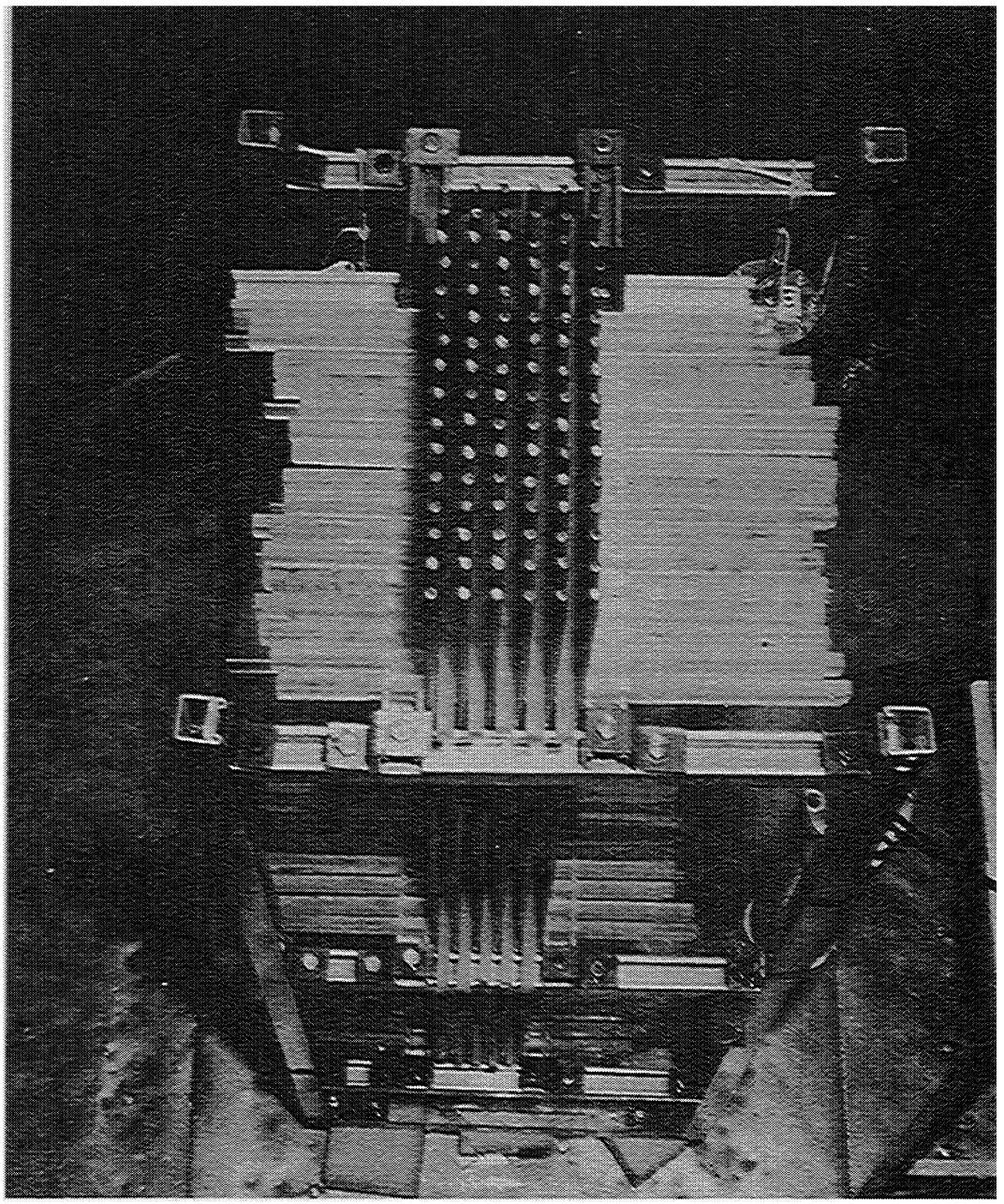


Fig. 5. A lattice of EBOR fuel pins assembled in square pattern. This lattice was critical when submerged.

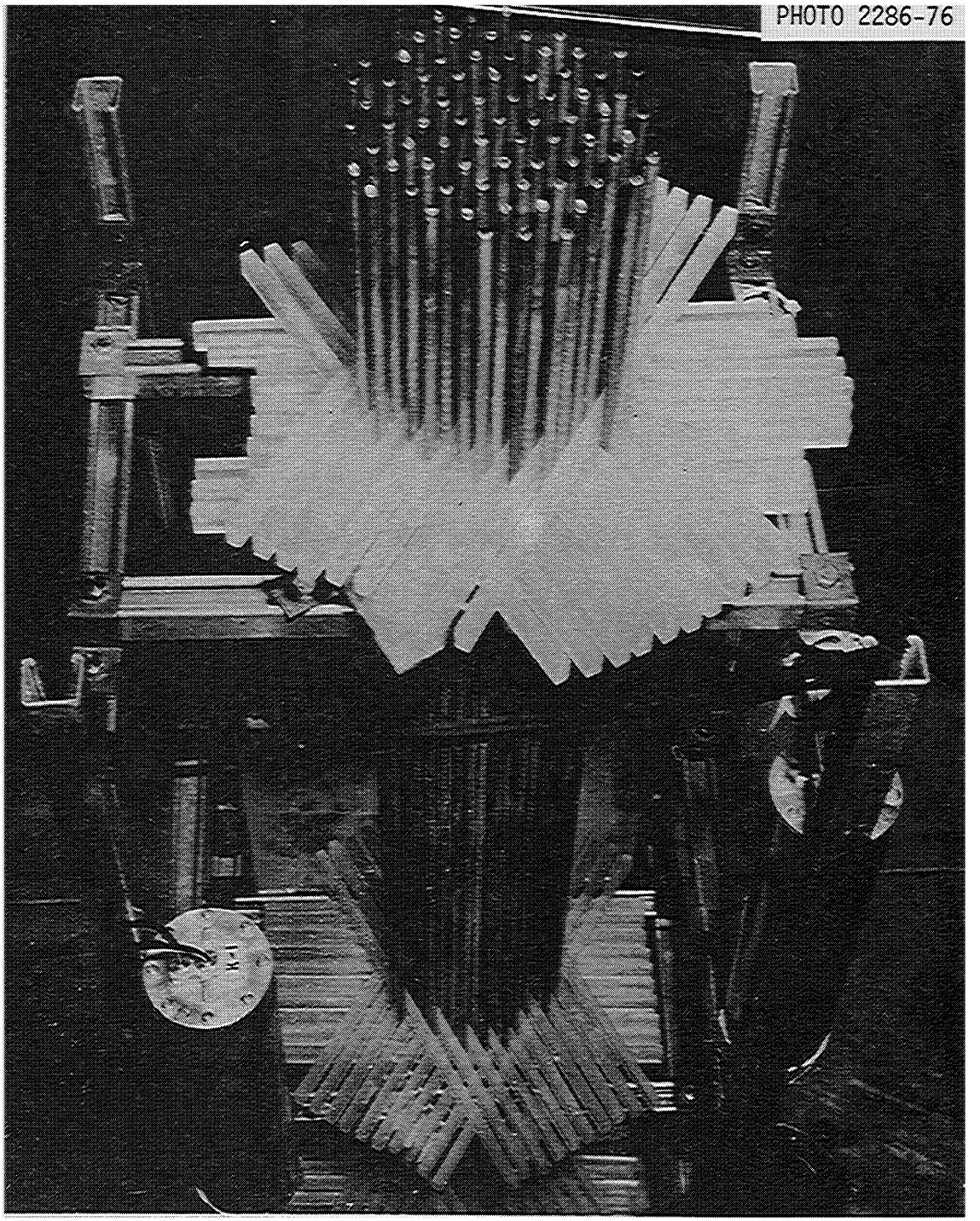



Fig. 6. A lattice of EBOR fuel pins assembled in triangular pattern. This lattice was critical when submerged.

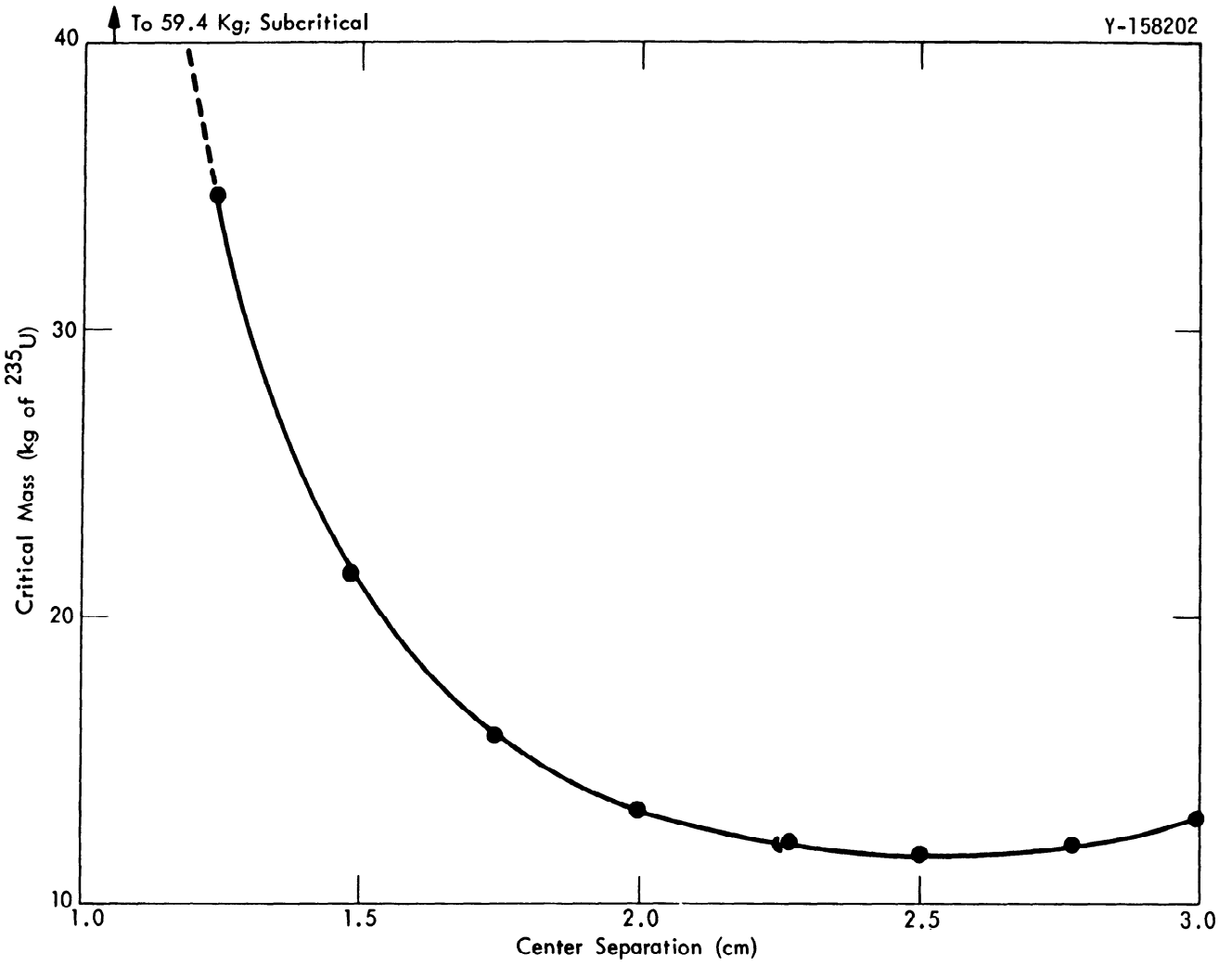



Fig. 7. The effect of separation between $U(62.4)O_2$-BeO fuel pins on the critical mass of water-reflected and -moderated lattices

## Table 3a. Lattices of EBOR fuel pins in water

| Surface Separation[a] (cm) | Surface Separation[a] (in.) | Center Separation[a] (cm) | Center Separation[a] (in.) | Critical Number of Pins | Critical Water Height Above Fuel (cm) | Critical Mass (kg of $^{235}U$) | Lattice Number[b] |
|---|---|---|---|---|---|---|---|
| 0.102 | 0.040 | 1.054 | 0.415 | (380)[c] | Subcritical | (59.37)[c] | 1 |
| 0.290 | 0.114 | 1.242 | 0.489 | 222[d,e] | (15.2)[d,e] | 34.68 | 2 |
| 0.536 | 0.211 | 1.488 | 0.586 | 138 | 30.8 | 21.56 | 3 |
| 0.790 | 0.311 | 1.742 | 0.686 | 102[f] | -21.3[f] | 15.94 | 4 |
| 1.046 | 0.412 | 1.999 | 0.787 | 85[d,g] | (15.2)[d,g] | 13.28 | 5 |
| 1.323 | 0.521 | 2.276 | 0.896 | 78[d,h] | (15.2)[d,h] | 12.19 | 6 |
| 1.300 | 0.512 | 2.253 | 0.887 | 77[f] | -3.9[f] | 12.03 | 7 |
| 1.554 | 0.612 | 2.507 | 0.987 | 75[d,i] | (15.2)[d,i] | 11.72 | 8 |
| 1.826 | 0.719 | 2.779 | 1.094 | 77[f] | -43.2[f] | 12.03 | 7 |
| 2.042 | 0.804 | 2.995 | 1.179 | 83[j] | -34.1[j] | 12.97 | 9 |
| 1.544[k] | 0.608[k] | 2.497[k] | 0.983[k] | 96[f] | -10.4[f] | 15.00 | 10 |
| 0.297 | 0.117 | 1.250 | 0.492 | 75[f] | -12.2[m] | 11.72 | 11 |

a. The reported separation is derived from the average of about 10 measurements of the outside dimensions of each lattice; the random orientation of the helix with relation to the spacers resulted in an effective average separation within a lattice, which could differ from the actual spacing of a pin by as much as twice the helix thickness.
b. See Table 3b for the actual lattice arrangement.
c. This lattice was sufficiently subcritical to preclude extrapolation to critical; the number and mass of fuel pins are those actually assembled.
d. The criticality of this lattice was postulated by bracketing the critical number of pins; one pin less than the number tabulated for the critical lattice resulted in subcriticality and one pin more produced criticality when the water height was below the top of the fuel.
e. A lattice of 223 pins was critical when the water was -50.3 cm.
f. This lattice was critical with the water level below the top of the fuel; removal of one pin resulted in subcriticality.
g. A lattice of 86 pins was critical when the water was -60.8 cm.
h. A lattice of 79 pins was critical when the water was -39 cm.
i. A lattice of 76 pins was critical when the water was -61.3 cm.
j. At this lattice spacing, 82 pins were slightly subcritical.
k. The average surface separation was 0.608 in. (1.544 cm) between pins in the 16-pin direction and 0.624 in. (1.585 cm) in the 6-pin direction.
m. The reactivity of this lattice with an effectively infinite top water reflector was 11 cents.

Table 3b. Lattices of EBOR fuel pins in water

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There is no significance to the spacing between symbols designating fuel pins in these diagrams. See Table 3a for the actual spacings.

minimum occurred at a surface separation of about 0.6 in., is inconsistent with the difference in spacing. Lattice Number 6 was essentially square in cross section, with three corner positions vacant, while Lattice Number 7 was "rounded"; this latter shape should result in less mass of fissile material required for criticality, assuming all else remained constant. Of the two known variables between these two lattices, it is apparent that the effect of array shape was dominant.

One of the objectives of the program was to establish the dimensions of a slab-shaped lattice of minimum thickness in which the pin separation was near that requiring minimum mass for criticality. Lattice Number 10 of Table 3, a 6 x 16-pin array shown in Fig. 5, was critical with the water about 10 cm below the top of the fuel; removal of one pin from a corner position resulted in subcriticality. A lattice 5 x 21 pins in cross section at the same spacing (nominally 0.6 in.) had no observed neutron multiplication. The dimensions of the critical slab-shaped lattice served as the basis for the experiments in which the fuel pins were made critical in solutions containing a neutron absorber and a uranium salt.

## Fuel Pins in Solution

The effect of the dissolver solution on the criticality of a slab-shaped lattice of this fuel was investigated at the lattice spacing that resulted in the minimum critical mass in the water-reflected and -moderated lattices. This lattice spacing had been determined to be about 0.6 in. surface separation.

In order to approximate the projected dissolver dimensions, the experiments with boron and with uranyl nitrate in the reflector-moderator water made use of a 20-in.-diam (50.8-cm-diam) stainless steel cylinder to which the solutions could be added by remote operation. This cylinder was mounted in the 9-ft-diam tank in order that it could be reflected by water. The pins were positioned by three Plexiglas grid plates, each 0.5 in. (1.3 cm) thick, that fit inside the solution cylinder and that were joined together by threaded steel rods. Holes drilled in these

plates at a pitch of 2.48 cm would allow a 7 x 19-pin lattice as shown in Fig. 8. The pins were supported by a 1-in.-thick Plexiglas pseudodisc that was raised 6 in. (15.2 cm) above the bottom of the cylinder by a 12-in.-OD Plexiglas tube with 1-in.-thick wall, notched to allow water under the base support. Because of the length of the fuel pins and the use of supports of minimal number and thickness, it was necessary that additions to a lattice be made with the assembly removed from the cylinder in order to assure the intended spacing over the entire length of the pins. Figure 9 is a photograph of this grid plate-support structure with fuel pins in position. Figure 10 shows the top of one of the lattices and of the support structure in the solution cylinder. Figure 11 gives the relevant dimensions of the entire structure as installed for the experiments.

As recorded in Table 4, the critical number of fuel pins in a water-moderated and -reflected slab mounted as described was 99. This number was increased to 114 upon substituting, for the water, an aqueous solution of $H_3BO_3$ at a concentration of 0.039 g of boron per liter and to 133 when the boron concentration was increased to 0.190 g/liter. Replacing the boron solution with $U(92.6)O_2(NO_3)_2$ dissolved in water at a $^{235}U$ concentration of 3.68 g/liter decreased the critical number of pins to 83. Subsequent addition of 0.315 g of boron to each liter of the uranyl nitrate solution brought the critical number back to 133.

In summary, reflection and moderation of the slab lattice by an aqueous boric acid solution containing 0.190 g of boron/liter increased the critical loading by 34% over the value with water alone. Substitution of uranyl nitrate solution containing 3.68 g of $^{235}U$/liter decreased the critical number of pins to 84% of that with water only. Subsequent addition of 0.315 g of boron/liter to that solution, however, increased the critical number 60%.

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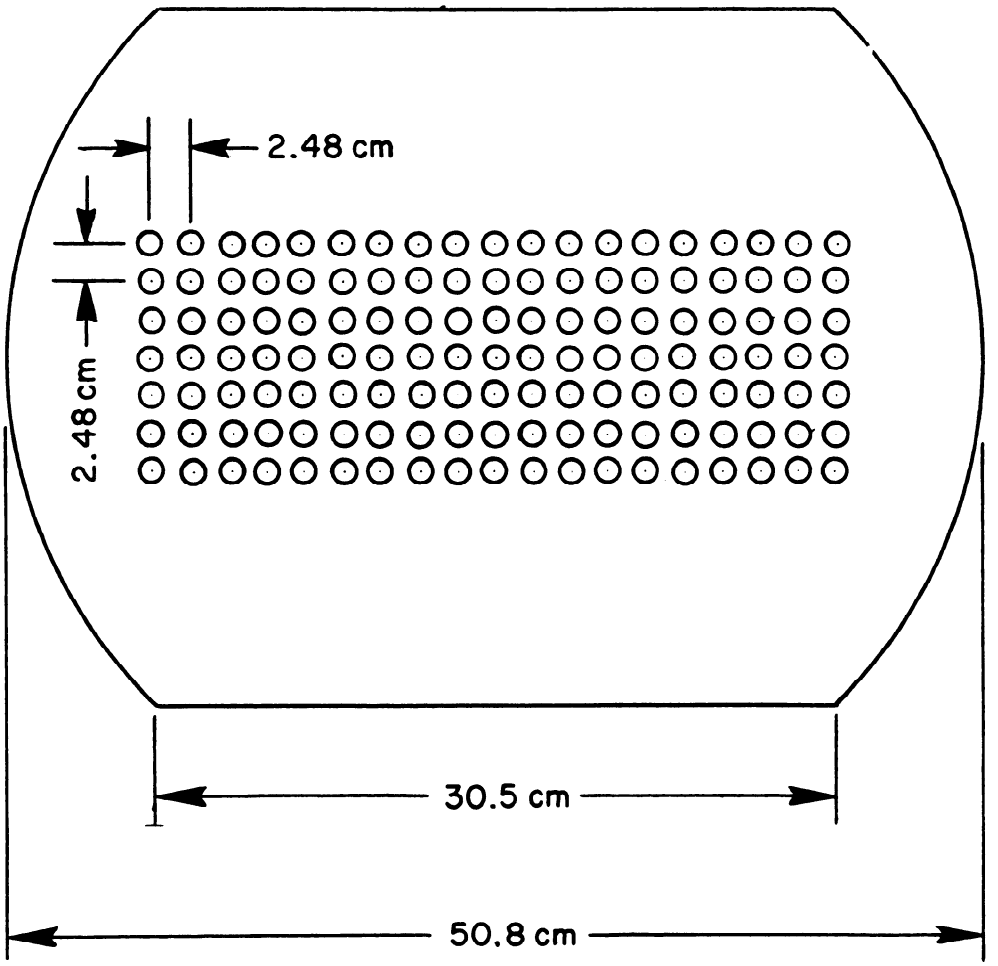



Fig. 8. Diagram of the Plexiglas grid plate for the solution cylinder

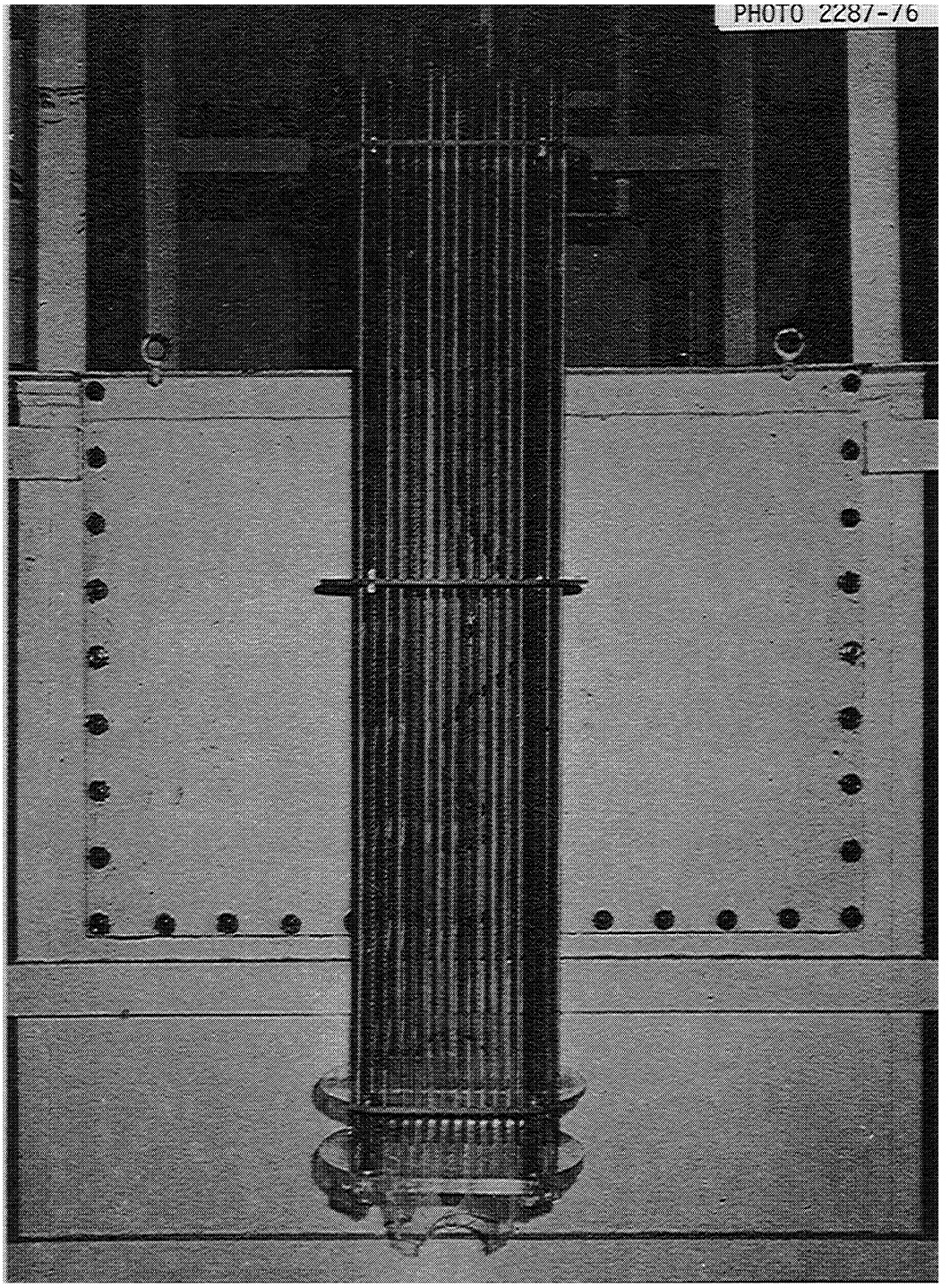



Fig. 9. Fuel pins in the support structure prior to installation in the solution cylinder

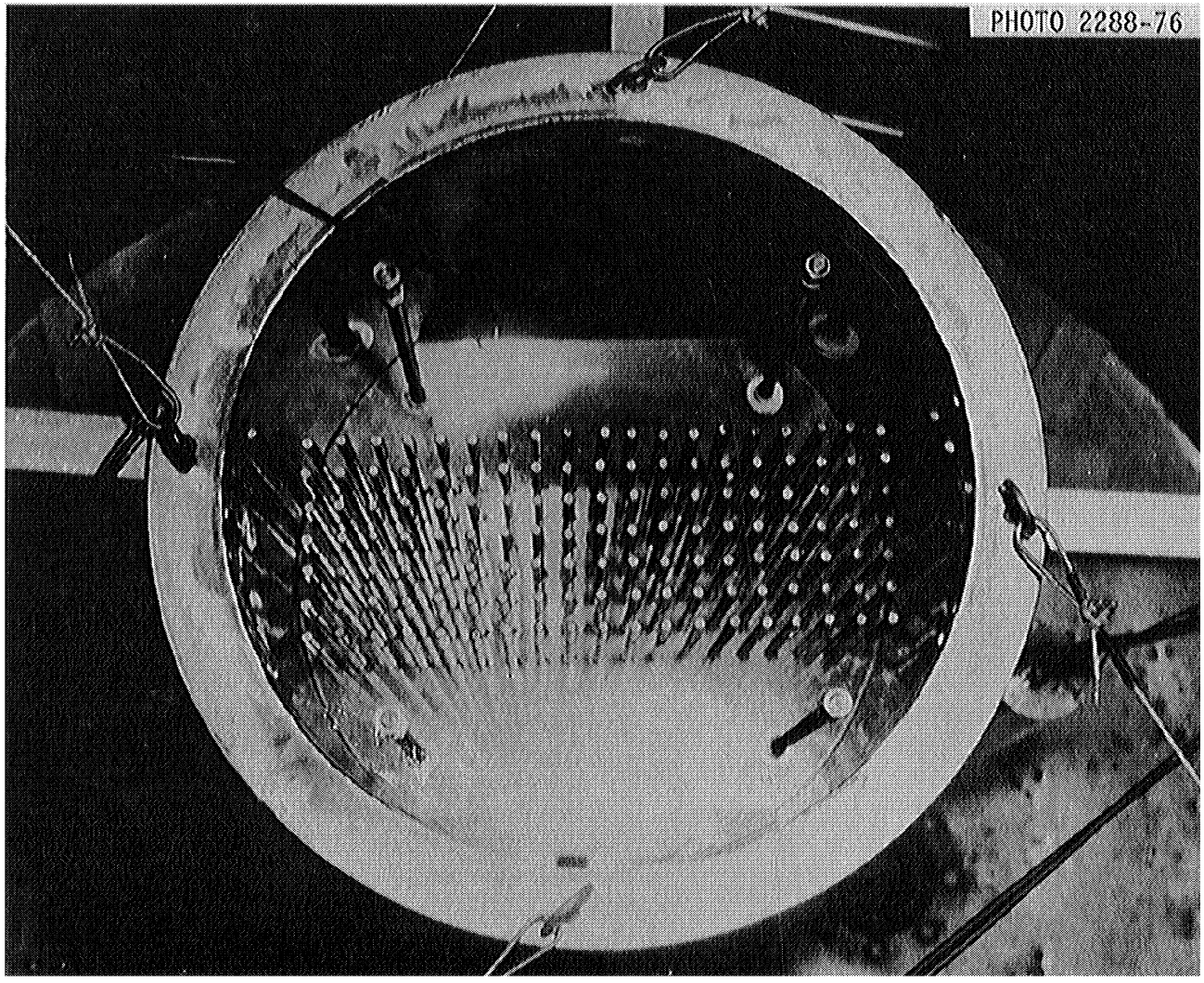



Fig. 10. Top view of a lattice in the solution cylinder

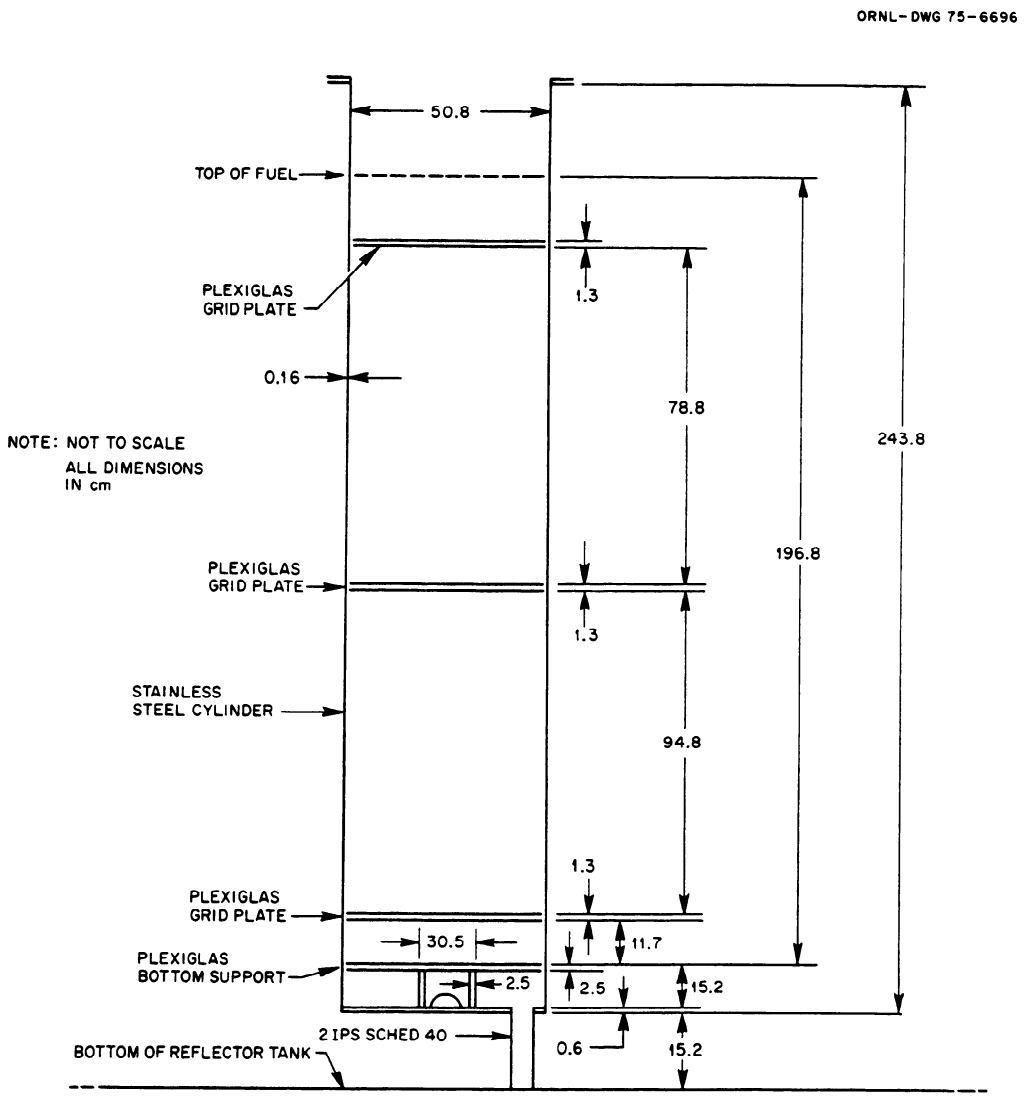


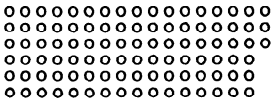
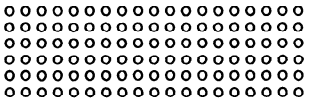
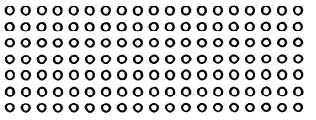
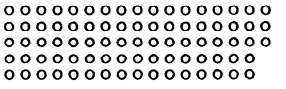

Fig. 11. Schematic of the cylinder and supports for fuel pins in aqueous solutions

Table 4. Slab lattices of EBOR fuel pins in aqueous solutions

| Aqueous Moderator/Reflector Composition | Concentration[a] of Solute (g/liter) | Number of Fuel Pins[b] | Mass of $^{235}U$ in Pins (kg) | Excess Reactivity of Submerged Lattice (cents) | Lattice Arrangement[c] |
|---|---|---|---|---|---|
| Water | -- | 99 | 15.47 | 1 | 1 |
| $H_3BO_3$ | 0.039 | 114 | 17.81 | 12 | 2 |
| | | 113 | 17.66 | 0 | 3[d] |
| $H_3BO_3$ | 0.190 | 133 | 20.78 | 0 | 4 |
| $U(92.6)O_2(NO_3)_2$ | 3.68 | 83 | 12.97 | 1 | 5 |
| $U(92.6)O_2(NO_3)_2$ + $H_3BO_3$ | 3.68<br>0.315 | 133 | 20.78 | 9 | 4 |

a. The values of concentration refer to either boron or $^{235}U$, as appropriate, in aqueous solution.

b. The grid plates spacing the slab lattices were mounted in a 50.8-cm-diam (20-in.-diam) stainless steel cylinder to which the solutions were added. This cylinder was reflected by an effectively infinite thickness of water on the bottom and sides to its full height. The bottom of the fuel pins was 21 cm (6 in.) above the bottom of the cylinder. The center spacing of the holes in the grid plates into which the fuel pins were inserted was 2.48 cm (0.975 in.).

c. 1.  2.  4.  5. 

d. Lattice Number 3 was produced by removing a corner pin from Lattice Number 2.

## ACKNOWLEDGEMENTS

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